

## 4 Task II: Solar Chemistry Research

Operating Agent:

Anton Meier, PSI, Switzerland (until July 31, 2016)

Philipp Furler, ETHZ, Switzerland (from August 1, 2016)

National Coordinators:

- Robbie McNaughton, CSIRO, Australia
- Liejin Guo, Xi'an Jiaotong University, China
- Gilles Flamant, CNRS-PROMES, France
- Karl-Heinz Funken, DLR, Germany
- George Karagiannakis, CERTH, Greece
- Alberto Giaconia, ENEA, Italy
- Yong-Heack Kang, KIER, Korea
- Claudio Estrada, UNAM, Mexico
- Jan van Ravenswaay, NWU, South Africa
- Alfonso Vidal, CIEMAT, Spain
- Anton Meier, PSI, Switzerland (until July 31, 2016)
- Philipp Furler, ETHZ, Switzerland (from August 1, 2016)
- Nathan Siegel, Bucknell University, USA

### 4.1 Nature of Work & Objectives

The primary objective of Task II—Solar Chemistry R&D—is to develop and optimize solar-driven thermochemical processes for the production of fuels and materials, and to demonstrate—at industrial scale—their technical and economic feasibility. The revised Task II structure for the current Term (2012-2016) is based on dedicated Activities to reflect the future challenges and to foster active participation of SolarPACES member countries:

- *Activity 1: Solar Fuels (SF)*
  - Demonstrate—at pilot scale—the most advanced processes for the solar production of synthetic fuels (e.g., cracking or steam reforming of NG, gasification of carbonaceous materials, carbo-thermal reduction of metal oxides).
  - Demonstrate medium and high temperature steam electrolysis and multi-step thermochemical processes (e.g., sulfur-based cycles) for the solar production of hydrogen.
  - Scale up the solar reactor technology of promising two-step  $H_2O/CO_2$ -splitting processes (e.g., Zn/ZnO & ferrite cycles) for the production of hydrogen, syngas, and liquid fuels.
- *Activity 2: Solar Materials (SM)*
  - Process chemical commodities using concentrated sunlight (e.g., aluminum, magnesium, silicon, zinc; lime and cement).
- *Activity 3: Thermo-chemical Storage (TS)*
  - Develop novel thermochemical storage systems for parabolic troughs and central receivers.
- *Activity 4: Technology Innovation (TI)*
  - Engineer innovative and cost-efficient high-temperature solar reactor materials and components.

- Design advanced heat recovery systems.
- Implement advanced concentrating optics for high solar flux intensities and high temperatures.
- *Activity 5: Research Infrastructure (RI)*
  - Optimize dedicated concentrating solar research infrastructure for large-scale testing of high-temperature thermochemical processes.
- *Activity 6: Market Penetration (MP)*
  - Enhance industry involvement in solar fuels production and promote market penetration of the most mature solar fuels technologies.
  - Interact with industry, government, and academia to develop country-specific “roadmaps to solar fuels” (Task II special activity).

### 4.2 Organization and Structure

The Task II Operating Agent (OA), PSI, Switzerland (until July 31, 2016) and ETHZ, Switzerland (from August 1, 2016), is responsible for organization, operation, and reporting. International solar chemical research, development and demonstration efforts are coordinated in cost, task and/or information-sharing activities by National Coordinators (NC), making use of an efficient network, for the rapid exchange of technical and scientific information. In 2015, we welcomed two new NCs: Liejin Guo from Xi'an Jiaotong University in China and George Karagiannakis from CERTH in Greece. In 2016, there was a change in the Task II management. Anton Meier from PSI in Switzerland was retired and resigned as Task II OA; Philipp Furler from ETH Zurich in Switzerland was elected as the new Task II OA.

The Task II Program of Work offers an up-to-date description of the national and international projects.

When appropriate, Task II conducts a status review on novel technologies for assessing their technical and economic feasibility. Task II is continuously striving to stimulate public awareness of the potential contribution of solar chemistry to clean, sustainable energy services. The Task II Annual Meeting provides a forum for presenting and discussing major technological achievements.

### 4.3 Status of Technology

This chapter provides a comprehensive overview of the many ways in which solar chemical technologies may be used for the delivery of clean, sustainable energy services. In 2014, special focus was on the solar thermal production of fuels (hydrogen and syngas) and chemicals for the power, transportation and chemical sectors of the world energy economy.

In 2015, solar chemistry research was presented at two major international conferences:

- *21<sup>st</sup> SolarPACES Conference*, Cape Town, South Africa, October 13-16, 2015; 12 oral presentations and 6 posters were submitted on solar fuels and chemical commodities.
- *ASME 2015 9<sup>th</sup> International Conference on Energy Sustainability*, San Diego, USA, June 28 – July 2, 2015; 13 technical presentations and 11 technical paper publications on “Solar thermochemical fuel production”.
- *ISES Solar World Congress*, Daegu, Korea, Nov 8-12, 2015; 5 oral presentations and 4 posters on solar chemistry research.

The 29<sup>th</sup> Task II Annual Meeting was held in conjunction with the 21<sup>st</sup> SolarPACES Conference, Cape Town, South Africa, October 12, 2015, attracting 28 attendees from 13 countries.

In the following, the most important achievements in 2015 of Task II related projects are summarized with updated information about project participation, objectives, status, and most relevant publications.

#### 4.3.1 ACTIVITY 1: SOLAR FUELS (SF)

##### Impact of Daily Startup-Shutdown Conditions on the Production of Solar Methanol over a Commercial Cu-ZnO-Al<sub>2</sub>O<sub>3</sub> Catalyst

Participant: ETHZ (CH)

Contacts: Aldo Steinfeld, [aldo.steinfeld@ethz.ch](mailto:aldo.steinfeld@ethz.ch)

Funding: ERC Advanced Grant (SUNFUELS)

Duration: January 1, 2014 – December 31, 2015

Background: The production of solar liquid fuels for transportation, such as synthetic diesel, kerosene, gasoline or methanol, requires the integration of solar syngas

production in a solar reactor with the subsequent catalytic gas-to-liquid conversion. Syngas storage or hybrid syngas production may be applied to ensure an uninterrupted feed during the night or periods of clouds, but their technical implementation is complex and their economics are uncertain. An alternative strategy is to operate the gas-to-liquid unit under a daily startup-shutdown regime to match the operation of the solar reactor.

Purpose: The objective is to investigate the effect of using syngas with the same composition as that obtained in a solar thermochemical reactor and of applying a daily startup-shutdown (DSS) routine matching the intermittent solar operation over a benchmark Cu-ZnO-Al<sub>2</sub>O<sub>3</sub> catalyst.

Achievements in 2015: The feasibility of the intermittent-daily startup-shutdown (DSS) production of methanol from a solar-like syngas mixture over a commercial catalyst was successfully demonstrated. The catalyst reached fast equilibration (10 h) in the presence of this syngas mixture and reversibly responded to changes in the concentrations of CO and CO<sub>2</sub> by mimicking fluctuations in the feed composition. Remarkably, its deactivation was even less pronounced over 27 cycles under DSS regime that for a corresponding time on stream under uninterrupted operation if the reactor was purged with H<sub>2</sub>-free syngas upon shutdown. Characterization and modeling indicated that this purging avoided the formation of inactive ZnCO<sub>3</sub> and minimized the oxidation of the Cu surface atoms.

Publications: [4.1]

##### Solar Dry (CO<sub>2</sub>) Reforming of CH<sub>4</sub> with an Isothermal Redox Membrane Reactor

Participant: ETHZ (CH)

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Funding: ERC Advanced Grant (SUNFUELS)

Duration: January 1, 2014 – December 31, 2015

Background: The dry (CO<sub>2</sub>) reforming of CH<sub>4</sub> (DRM) into syngas proceeds at elevated temperatures and is endothermic by about 247 kJ/mol according to the net reaction: CH<sub>4</sub> + CO<sub>2</sub> → 2CO + 2H<sub>2</sub>. Typically, catalyst deactivation by coke deposition from DRM side-reactions is alleviated by the use of rare noble metal catalysts. Such expensive materials are not required at above 900°C and, additionally, the deposited carbon can be oxidized with the lattice oxygen of a metal oxide redox material.

We have developed and demonstrated the feasibility of DRM with an isothermal redox membrane reactor that combines the benefits of a continuous isothermal solar fuel production with those of thermochemical DRM. The concept is illustrated in Figure 4.1. It is based on the continuous splitting of CO<sub>2</sub> to CO on one

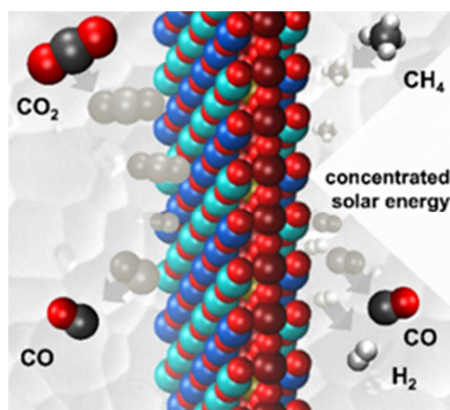


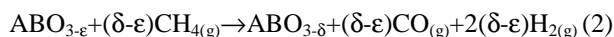
Figure 4.1.  $\text{CO}_2$  reforming of  $\text{CH}_4$  with an isothermal redox membrane reactor.

side of the membrane and the partial oxidation of  $\text{CH}_4$  with the lattice oxygen to syngas on the other side, with oxygen (vacancies) being conducted across the membrane.

Thus,  $\text{CO}_2$  is reduced by the oxygen vacancies of a perovskite membrane into CO:



The perovskite has  $\text{ABO}_{3-\delta}$  stoichiometry, where A and B are metal cations in twelve- and six-coordinated interstices, respectively, and  $\delta$  is the oxygen non-stoichiometry. The oxygen that is absorbed by the membrane on one side is conducted along an oxygen vacancy gradient to the other membrane side and abstracted from the solid via reforming of  $\text{CH}_4$  into CO and  $\text{H}_2$ :



Since oxygen is continuously replenished from  $\text{CO}_2$ , formation of a thick layer of reduced metal oxide is avoided, which, in turn, augments oxygen conduction kinetics. Overall,  $\text{CO}_2$  and  $\text{CH}_4$  are reformed into solar syngas, whose energetic value has been solar-upgraded by a factor of 1.31 over that of the methane feedstock. To achieve this, the redox membrane provides variable metal oxidation states accessible through oxidation with  $\text{CO}_2$  and reduction with  $\text{CH}_4$  at elevated temperatures.

**Achievements in 2015:** To demonstrate the feasibility of this concept, we have selected  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$  (LSCF) as the redox material. This is based on its superior oxygen conductivity and typically high oxygen vacancy concentrations, stability of the non-stoichiometric cubic phase, the redox activity of cobalt and iron, and the relatively low carbonate formation tendency. One-end open tubular LSCF redox membranes were fabricated via a phase inversion technique and evaluated for DRM using a tube-in-shell membrane reactor. Figure 4.2A shows the isothermal production rates of CO via  $\text{CO}_2$  splitting in the membrane lumen at steady-state. Figure 4.2B shows the equivalent rates for the production of CO and  $\text{H}_2$  via  $\text{CH}_4$  reforming in the reactor shell. The formation of CO from  $\text{CH}_4$  is relatively stable over a course of 20 min and, as expected from mass balance, approximately equal to CO formation

from  $\text{CO}_2$ . Purging the reactor in absence of the redox membrane at these temperatures with  $\text{CO}_2$  did not yield CO. This confirms that  $\text{CO}_2$  is reduced into CO at the inner membrane surface and that the abstracted oxygen is conducted across the membrane to activate  $\text{CH}_4$  at the outer membrane surface, yielding CO and  $\text{H}_2$ .

**Publications:** [4.2]

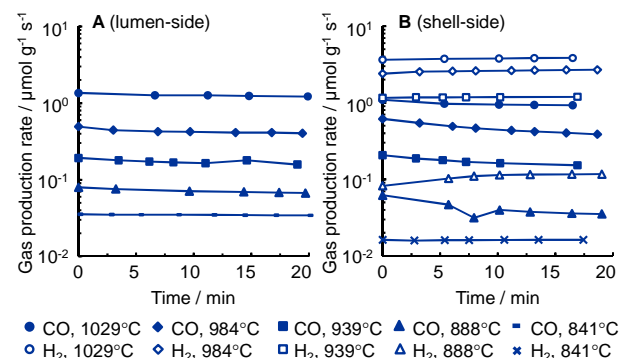


Figure 4.2. Isothermal steady-state production rates of (A) CO from 1 bar  $\text{CO}_2$  in the lumen of a tubular LSCF redox membrane and of (B) CO and  $\text{H}_2$  from 1 bar  $\text{CH}_4$  in the reactor shell. Solid lines are a guide only.

## Zn-Based Thermochemical Cycle for Splitting $\text{H}_2\text{O}$ and $\text{CO}_2$

**Participants:** PSI (CH), ETHZ (CH)

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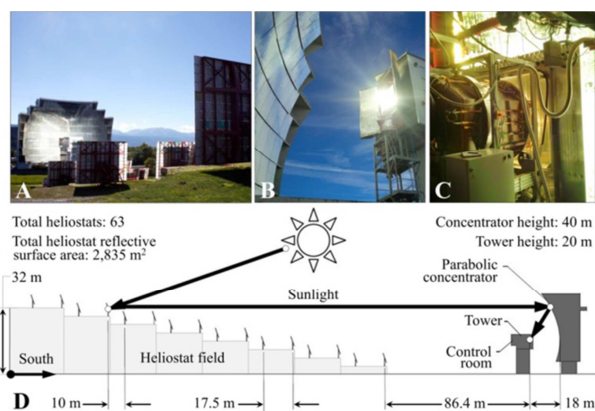
**Funding:** SFOE – Swiss Federal Office of Energy

**Duration:** January 1, 2003 – December 31, 2015

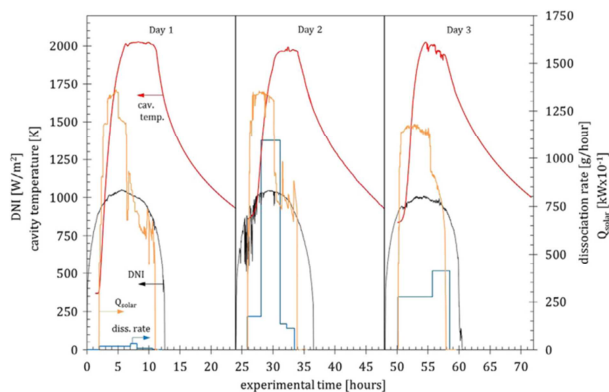
**Background:** The solar two-step Zn/ZnO redox cycle for splitting  $\text{H}_2\text{O}$  and/or  $\text{CO}_2$  inherently operates at high temperatures and utilizes the entire solar spectrum, and as such provides a thermodynamically favorable path to efficient solar fuel production. The first, endothermic step is the thermolysis of ZnO to Zn and  $\text{O}_2$  using concentrated solar radiation as the source of process heat. The second, non-solar, exothermic step is the reaction of Zn with mixtures of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  yielding high-quality syngas (mainly  $\text{H}_2$  and CO) and ZnO; the latter is recycled to the first solar step, resulting in the net reactions  $\text{CO}_2 = \text{CO} + 0.5\text{O}_2$  and  $\text{H}_2\text{O} = \text{H}_2 + 0.5\text{O}_2$ , respectively.

**Purpose:** The primary purpose of the current research is to optimize the solar reactor technology for the thermal dissociation of ZnO at laboratory scale (solar power input of 10 kW) and to successfully demonstrate and characterize the fully integrated reactor at pilot scale (solar power input of 100 kW). Conceptual designs and preliminary economics for commercial solar Zn production and storage facilities will be developed based on this characterization and implemented on large-scale concentrating solar power (CSP) tower technology.

**Achievements in 2015:** Successful demonstration and promising characterization of a solar reactor pilot plant for thermal reduction of ZnO as part of a two-step water and CO<sub>2</sub> splitting cycle has been accomplished at the 100 kW<sub>th</sub> scale in the 1 MW Solar Furnace (MWSF) of PROMES-CNRS in Odeillo, France (Figure 4.3). The solar reactor pilot plant was operated for over 97 h and achieved sustained reaction temperatures well above 2000 K, while demonstrating ZnO dissociation rates as high as 28 g/min totaling over 28 kg of processed reactant during 13 full days of experimentation [4.3]. In-situ, high temperature, flow visualization of the quartz window enabled the unimpeded operation of the solar reactor [4.4]. As many as three consecutive full day experiments were conducted without complication (Figure 4.4). Solar power delivered to the reaction cavity ranged between 90 and 128 kW<sub>th</sub>, at peak solar concentrations as high as 4671 kW/m<sup>2</sup>. The products Zn and O<sub>2</sub> were quenched with Ar(g) and recovered in a filter battery, where collected particles contained molar Zn-content as high as 44%. During experimentation, switching between product collection filter cartridges resulted in 54 unique experiments, where a maximum solar-to-chemical efficiency of 3% was recorded for the solar reactor. Robust characterization of the product quenching device revealed inherent limitations in its effectiveness, and thus solar-to-fuel energy conversion efficiency was limited to 0.24% if it would have been possible to supply 4640 L/min of Ar(g). Further, only a limitation on available experimental time prohibited the demonstration of significantly higher dissociation rates, achievable with higher ZnO reactant feed rates. While the use of large volumes of quenching Ar(g) to separate the reaction products remains a significant obstacle to achieving higher solar-to-fuel efficiencies, demonstration of solar reactor technology at the pilot-scale represents significant progress toward the realization of industrial-scale solar fuels production.



**Figure 4.3.** Schematic representation and images of the 1-MW solar furnace (A,B,D) and 100-kW<sub>th</sub> reactor installation (C). The parabolic concentrator is seen from the heliostat field during an experiment in (A), and from below the tower (B) where the reactor is installed. In (C) the reactor can be seen from behind, with the parabolic concentrator illuminated in the background [4.4].



**Figure 4.4.** Data plot for three days of consecutive experiments. DNI, cavity temperature, dissociation rate, and solar power input are plotted across 72 h of continuous data collection [4.3].

Recently, a techno-economic analysis of hydrogen production via the Zn/ZnO thermochemical cycle was performed using a scaling of the 100 kW<sub>th</sub> ZnO reduction reactor to a power level most likely necessary for economical operation. Results confirmed the significant impact of quench gas use: for a 110 MW<sub>th</sub> plant producing 4.19 tons per day of H<sub>2</sub>, a hydrogen price of \$8/kg was forecast without considering quenching gas, and over \$50/kg when considering it [4.5]. In contrast to other techno-economic evaluations<sup>1</sup>, where higher prices were assumed surmountable with appropriate policy incentives, Koepf *et al.* [4.6] conclude that one should not proceed with solar fuel production using the Zn/ZnO thermochemical cycle unless significant advances are made in product separation and/or efficient inert gas recycling.

**Publications:** [4.3]-[4.6]

## CCEM HyTech – Vacuum Carbothermic Reduction of ZnO

**Participants:** PSI (CH)

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**Funding:** CCEM – Swiss Center for Energy and Mobility

**Duration:** February 1, 2012 - June 30, 2015

**Background:** The application of vacuum has been proposed in order to reduce reaction temperatures for the carbothermal reduction of metal oxides. According to Le Chatelier's principle, decreasing the partial pressure of product gases shifts the thermodynamic equilibrium to lower temperatures. While application of vacuum implies an energy penalty due to pumping to lower pressures, it is expected that the benefits of vacuum

<sup>1</sup> Nicodemus J.H., McGuinness M., Maharjan R. (2014). A Thermodynamic and Cost Analysis of Solar Syngas From the Zn/ZnO Cycle. *J. Solar Energy Eng.*, 137, 11012.



operation will outweigh these costs. The benefits of a lower reaction temperature include less stringent material constraints for reactor construction, and an overall positive impact on the economic viability of the reduction process as less concentrated solar power is required to drive the reaction.

**Purpose:** The objective is to develop a solar-driven vacuum aerosol reactor concept for the carbothermal reduction of zinc oxide and other metal oxides.

**Achievements in 2015:** A solar-driven aerosolized particle reactor under vacuum was tested for carbothermal reduction of zinc oxide using concentrated solar power [4.7]. The reactor concept is based on the downward flow of zinc oxide and carbon particles, which are indirectly heated by an opaque intermediate solar absorption tube (Figure 4.5). The particles are rapidly heated to reaction temperature and reduced within residence times of less than 1 s. In the continuous feeding experiments, maximum sustained temperatures close to 2000 K and heating rates as fast as  $1400 \text{ K min}^{-1}$  could be achieved for pressures between 1 and 1000 mbar. Reactant conversions of up to 44% were obtained at 1000 mbar. It was found that a reduction in system pressure leads to a decreased particle residence time (as low as 0.09 s), and therefore low conversion (as low as 1%), thus partially diminishing the positive thermodynamic effects of vacuum operation. Experimental results validate the robust and versatile reactor concept, and simultaneously highlight the necessity of balancing the system design in order to optimize the conflicting influence of vacuum operation and reacting particle residence time.

Ultrasonic vibratory and rotary valve particle feeders have been designed, constructed, and investigated for application to feeding reactant powder to a solar thermochemical drop-tube reactor [4.8]. Zinc oxide and carbon particles are fed continuously to the drop-tube under vacuum pressures as low as 1 mbar. The particles are probed in situ by laser transmission measurements with the aim to characterize particle residence time, axial and radial dispersion as a function of operating pressure. The ultrasonic feeder disperses particles well and can be operated at mass flow rates in the range of

$57\text{--}288 \text{ mg min}^{-1}$ . The rotary valve feeder operates in the mass flow range of  $3.46\text{--}41.96 \text{ g min}^{-1}$  and exhibits reduced particle dispersion due to discrete pulsing mass flow created from the rotating valve. The time resolved transmission signals reflect characteristic changes under different experimental vacuum conditions (Figure 4.6).

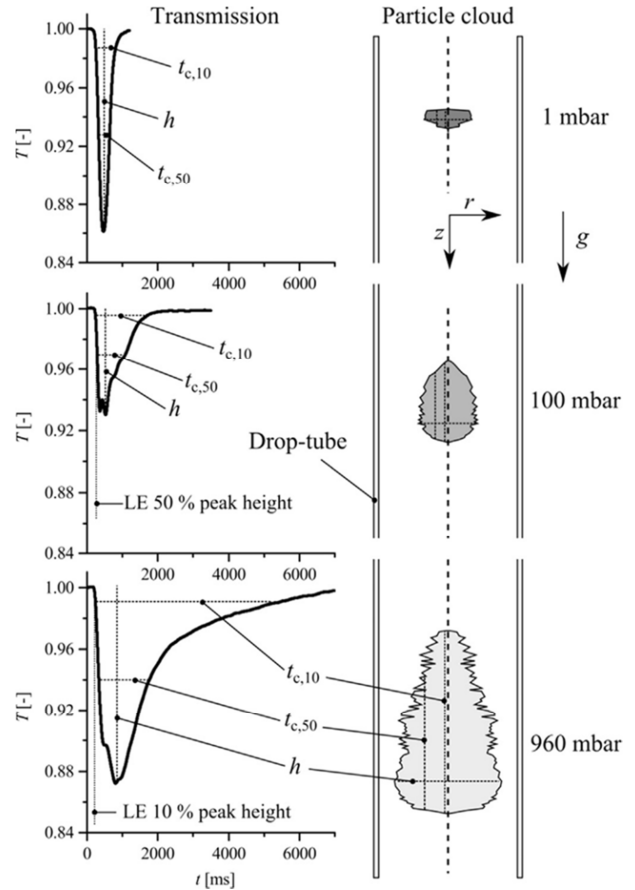


Figure 4.6. Conceptual illustration of the correlation between average peak shape characteristics of the transmission data and corresponding average particle cloud density at system pressures between 1 and 960 mbar [4.8].

Particles traveling through the measurement zone at 1 mbar exhibit residence and clearance times of 0.05 s and 0.52 s, respectively. At 960 mbar, residence and clearance times are increased to as much as 0.16 s and 3.98 s, respectively. Particles falling at 1 mbar show radial dispersion three times less than those falling under ambient pressure. A critical result of the functional characterization of powder feeding under vacuum is a potential reaction capacity limitation at low vacuum pressures due to short particle residence time and narrow axial dispersion.

**Publications:** [4.7]–[4.8]

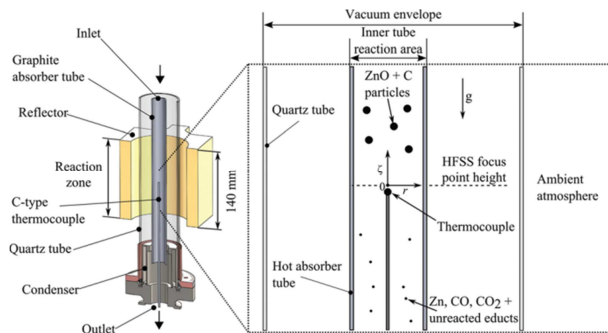


Figure 4.5. Schematic of the vacuum aerosol reactor as a cross-sectional view, and an enlarged view of the graphite absorber tube showing the hot reaction zone and particle flow. The graphite tube radial and vertical axes are denoted by  $r$  and  $z$ , respectively [4.7].

## SolarSynGas – Detailed Understanding of Reactions and Processes for Solar Thermochemical H<sub>2</sub>O and CO<sub>2</sub> Splitting

**Participants:** DLR (D), KIT (D), TU Clausthal (D), ETH (CH)

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**Funding:** EC funded project, cost shared: € 4,400,000

**Duration:** October 1, 2012 – Sep. 30, 2017

**Background:** The solar thermal dissociation of H<sub>2</sub>O or CO<sub>2</sub>, respectively, directly combines low economic value of its reactants with high economic value of its products and therefore comes in for great commercial interest. It represents a technology free of greenhouse gas emissions and fossil fuels consumption. The challenge is to couple solar energy as the driving energy for the reaction. In order to lower the extremely high operating temperatures required for direct dissociation of H<sub>2</sub>O and CO<sub>2</sub> and to eliminate the need for high-temperature gas separation, current research concentrates on the so-called thermo-chemical H<sub>2</sub>O/CO<sub>2</sub>-splitting cycles. Such “multistep” processes to decompose H<sub>2</sub>O/CO<sub>2</sub> into H<sub>2</sub>/CO and oxygen via two or more chemical reactions require much lower temperatures than those needed for direct thermolysis. The German Helmholtz Association has set up a Virtual Institute on this topic to understand and use fundamental mechanisms of the redox reactions to form a basis for the synthesis of metal oxide redox materials with improved efficiency and lifetime and to understand the influence of key process parameters.

**Purpose:** Main objective is the systematic development of the components necessary for solar fuel generation from water and carbon dioxide: investigation of the processes and materials involved and implementation into solar powered reactor technology. Optimized functional ceramics will be developed as redox materials for solar H<sub>2</sub>O and CO<sub>2</sub> splitting. Basic understanding of interrelations between atomic structure and transport, microstructure, reactivity and life time of the materials will be developed to design optimized reactor and process concepts for implementation.

**Achievements in 2015:** The project had to undergo an Intermediate Evaluation in 2015. As a result, the Virtual Institute was prolonged until the end of 2017 with an intensified focus on investigating detailed mechanisms of the reactions and the transport processes and on efficient heat recovery [4.9]–[4.11].

Developing an efficient redox material is crucial for thermochemical cycles that produce solar fuels, enabling a sustainable energy supply (Figure 4.7). The effects of varying the rare-earth (RE) content  $y$  on the fuel productivity and long-term stability were investigated for  $\text{Ce}_{0.85-y}\text{Zr}_{0.15}\text{RE}_y\text{O}_{2-0.5y}$  with RE = Y, La, Sm and Gd [4.12]. Compared to the none-RE-doped reference material,  $\text{Ce}_{0.85}\text{Zr}_{0.15}\text{O}_2$ , none of the compositions exhibits higher performances. However, long-term cy-

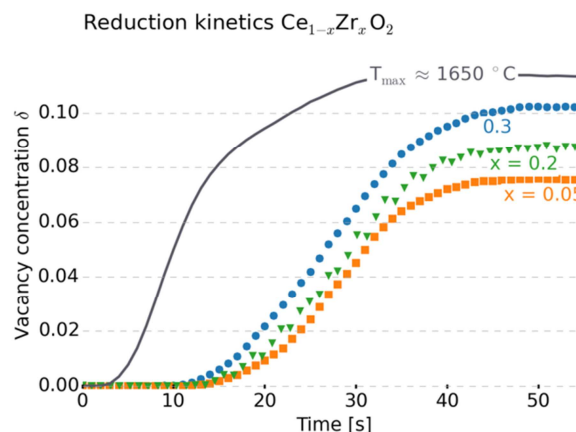


Figure 4.7. Oxidation and reduction reaction kinetics of mixed cerium zirconium oxides [4.13].

cling of more than 80 cycles reveals enhanced performance due to rare-earth doping.  $\text{Ce}_{0.85}\text{Zr}_{0.15}\text{O}_2$  suffers from linear degradation of the yields and of the CO:O<sub>2</sub> ratio, which is attributed to declining oxidation kinetics, whereas for instance  $\text{Ce}_{0.82}\text{Zr}_{0.15}\text{Sm}_{0.03}\text{O}_{1.99}$  features stable yields and kinetics. The suggested rationale behind is found in a vacancy-depleted region that occurs in the grains of  $\text{Ce}_{0.85}\text{Zr}_{0.15}\text{O}_2$ . While cycling the specific surface decreases and the impact of these regions on the reaction rate increases, which leads to declining oxidation kinetics. In contrast,  $\text{Ce}_{0.82}\text{Zr}_{0.15}\text{Sm}_{0.03}\text{O}_{1.99}$  displays structural vacancies corresponding to the  $\text{Sm}^{3+}$ , which remain during oxidation. Because of these structural vacancies, the oxygen bulk transport is enhanced, resulting in a constant reaction rate. Because of the long-term cycling, rare-earth doping is beneficial particularly for the oxidation kinetics and, hence, important for the technical realization of the process.

A new concept for heat recovery from redox particles in a high temperature solar thermochemical process has been proposed that uses ceramic spheres as solid heat transfer medium (Figure 4.8). This concept can be

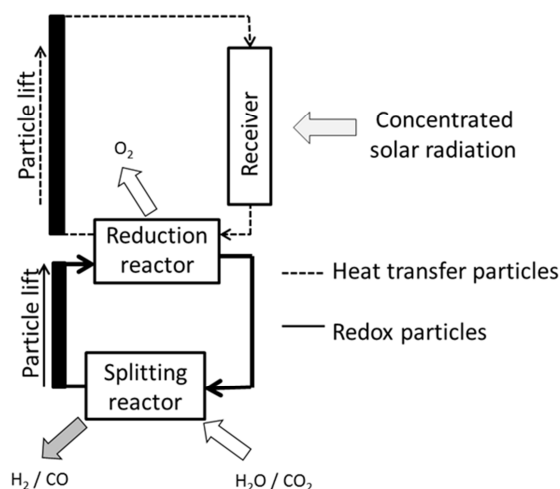


Figure 4.8. Process concept applying to redox material particles.

implemented into any particle reactor and offers sufficiently high heat recovery rates and thus high overall system efficiencies. A detailed model to calculate the performance of the concept in consideration of temperature dependent material data and several other influencing factors is described. An analysis of the diameters of the redox particles and the heat transfer spheres is carried out. The performance is investigated for alumina as the heat transfer medium and ceria as the redox material. It is found that the molar flow ration needs to be optimized regarding the contact time and the heat recovery rate only increases slightly over a contact time of  $t = 10$  s. The system reaches a heat recovery rate over 70% in case of six stages, connected in a quasi-counter-current principle. Further investigations shall address the transient operational conditions of a solar thermochemical plant. In addition, we shall measure the heat transfer coefficient between heat transfer spheres and redox particles.

Publications: [4.9]-[4.13]

## SOL2HY2 – Solar-to-Hydrogen Hybrid Cycles

Participant: EnginSoft (I), Aalto (FIN), DLR (D), ENEA (I), Outotec (FIN), Erbicol (CH), Woikoski (FIN)

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Funding: EC (FCH JU) contribution: 1,991,115 € (cost shared)

Duration: June 1, 2013 – November 30, 2016

Background: The Hybrid Sulfur Cycle (HyS) for solar hydrogen production is considered as one of the most promising thermochemical cycles in terms of future realization at industrial scale. This water splitting process generates hydrogen and oxygen using thermal energy and consists of two stages. In the first reaction step, sulfuric acid is decomposed at 800–1200 °C to form sulfur dioxide (SO<sub>2</sub>) and oxygen; the latter is separated as a by-product. In the second reaction step, sulfur dioxide is electrolyzed with water at temperatures below 100 °C to produce hydrogen and fresh sulfuric acid, which is recycled to the first reaction. The power demand of this sulfur dioxide depolarized electrolysis (SDE) is only about one-tenth of the demand of conventional water electrolysis, so that the overall efficiency for the production of hydrogen can be significantly increased.

In a variant of the HyS – the so-called Outotec Open Cycle (OOC) – SO<sub>2</sub> is taken as a waste product of industrial processes (e.g. metallurgical industry) and converted by SDE into hydrogen and sulfuric acid. Combining the OOC process with existing sulfuric acid plants is considered to facilitate the market entry of the SDE technology as a first step for realization of the HyS process.

Purpose: First, plant concepts are drafted combining HyS and OOC for most efficient and economic hydrogen production during on-sun and off-sun operation. Subsequently, flowsheets are developed to simulate the process [4.14] forming the basis of a techno-economic study. Furthermore, the project is focused on the development of prototypes and system models for key components of the process (i.e. solar sulfuric acid cracker (SAC) and SDE). DLR develops a SAC demonstration plant on its solar tower in Juelich, Germany, supported by ENEA preparing the catalyst for the decomposition reaction and Erbicol providing SiC foam structures used as solar absorber. A SDE prototype is developed by Aalto preparing a stack for laboratory testing. Moreover, balance of plant (BoP) components are analyzed with particular focus on the SO<sub>2</sub>/O<sub>2</sub> gas separation unit. In addition, a software tool for multi-objective optimization is implemented by EnginSoft combining process flowsheets and system models developed in the project. The consortium is completed by the industrial partners Outotec (Technology Company in the metallurgy and sulfuric acid industry) and Woikoski (Gas Company for hydrogen production and distribution).

Achievements in 2015: Active flowsheets for different plant concepts were prepared with variable “openness” meaning the amount of waste SO<sub>2</sub> being consumed producing hydrogen and excess sulfuric acid. This allows studying the two extreme cases being (1) the HyS with no consumption of waste SO<sub>2</sub> and (2) the OOC requiring no solar thermal power for sulfuric acid recycling, as well as all combinations of the two process variants. In addition, the influence of all relevant process parameters including plant size and location can be analyzed with the developed flowsheets. The results of this process simulation are the input for a custom made software tool for multi-objective optimization, which was implemented to determine the optimal plant configuration.

The SAC demonstration plant was assembled on DLR’s solar tower in Juelich, Germany and initially operated in on-sun tests with air and water in autumn 2015 (Figure 4.9) to determine the thermal characteristics of the system. An outlet gas temperature of 1000 °C suitable for sulfuric acid cracking was achieved at an absorber front temperature of 1200 °C and a solar power on the aperture of 50 kW. After modification of the demo plant during the winter months, the on-sun tests are continued in spring 2016. The available system model of the solar receiver [4.15] was compared to

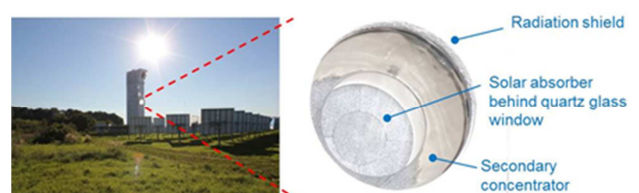


Figure 4.9. Solar receiver of demonstration plant for sulfuric acid cracking (right) during initial operation in 2015 on research platform of DLR’s solar tower in Juelich, Germany (left).

experimental data of the initial operation showing good agreement between simulation and experiment. An iron(III)oxide catalyst was developed for the SAC and qualified in laboratory test deriving a kinetic model.

With the developed prototype SDE stack extended laboratory tests were carried evaluating coating materials for the bipolar plates [4.16]. SO<sub>2</sub> carry-over and sulfur formation were determined as the two main reasons for drop of the stack current during operation [4.17]. In addition, the existing SDE working model was improved and validated with experimental data.

Publications: [4.14]-[4.17]

## **PECDEMO – Photoelectrochemical Demonstrator Device for Solar Hydrogen Generation**

Participants: HZB (D), EPFL (CH), IIT (IL), DLR (D), UPORTO (PT), EVONIK INDUSTRIES AG (D), SOLARONIX SA (CH)

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Funding: Europe's Fuel Cell and Hydrogen Joint Undertaking (FCH JU) co-funded project, cost shared: € 3,394,010 (Grant Agreement No. 621252)

Duration: April 1, 2014 – March 31, 2017

Background: The evolving exploitation of renewable energy involves the need of large-scale energy storage. A promising route with low carbon emissions is the generation of hydrogen using hybrid photoelectrochemical-photovoltaic (PEC-PV) tandem devices to split water. The tandem design approaches an optimal utilization of the solar input.

Purpose: The main objective of this project is the demonstration of a stand-alone solar water splitting device with an active area of at least 50 cm<sup>2</sup> and a solar-to-hydrogen efficiency of 8% or more which shows stability for >1000 h. Special attention is turned on scalability, the use of earth-abundant materials, and suitable large-area technologies. Extensive techno-economic and life-cycle analyses form part of the work plan. The scheduled contributions of DLR comprise investigations and optimization work with respect to the combination of PEC-PV devices with solar concentrators, experimental assessment of PEC-PV devices employing DLR's two-axis tracking solar concentrator and test platform SoCRatus (Solar Concentrator with a Rectangular Flat Focus), and techno-economic analyses.

Achievements in 2015: DLR developed and evaluated different concentrator concepts regarding the technical feasibility and suitability to irradiate PEC-PV devices. A medium range of concentration ratios from 20-40 was considered. Further requirements were an appropriate uniformity of irradiation on the active area and an efficient use of the solar spectrum. Three different hydro-

gen production scenarios were introduced: Hydrogen refueling station, an industrial process, and a single home application with an average capacity of 400 kg/day, 4000 kg/day, and 1 kg/day, respectively. Seville (Spain) and the Negev (Israel) featuring high levels of global solar irradiation were chosen as promising locations for pilot plant sites for further analyses. The sizes of the collector fields corresponding to the active PEC-PV areas operating under one sun condition were calculated for the three scenarios and the two locations. The sizes range from 89.1 m<sup>2</sup> to 378,139 m<sup>2</sup> and thus cover a broad range of conceivable commercial interest. Initial process designs were compiled and analyzed regarding flows of mass and energy in the design point. Input data for a first life-cycle analysis was compiled and a preliminary economic analysis was conducted, particularly addressing scale-up and solar concentration.

Publication: [4.18]

## **DuaSol – Dual Utilization of Solar Energy: Generation of Hydrogen in Waste Water Treatment**

Participants: H.C. Starck GmbH (D), JLU (D), LUH (D), HSU (D), DLR (D)

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Funding: Federal Ministry of Education and Research of Germany co-funded project, cost shared: € 3,437,076 (03SF0482B)

Duration: September 1, 2014 – August 31, 2017

Background: Hydrogen generation employing photocatalytic or photoelectrochemical (PEC) processes allows a carbon free storage of intermittent solar energy in a promising energy carrier. However, economically feasible systems have not yet been introduced.

Purpose: The main objective of this project is the development of a system which stores solar energy by generating hydrogen as well as degrades organic pollutants in waste water in an economically viable and industrially applicable manner. For this purpose, a photoelectrochemical (PEC) tandem device with cold sprayed photoelectrodes for simultaneous production of hydrogen and treatment of waste water will be designed and constructed. The work plan comprises (1) the development and tests of enhanced photocatalysts, (2) the adaption of morphology and identification of suitable production routes, (3) mechanistic investigations of photocatalysts and electrodes, (4) the production of photoelectrodes by cold gas spraying, and the development of solar reactors. The scheduled contributions of DLR encompass the development and evaluation of different reactor concepts, the construction of scalable solar test reactors, the experimental assessment of different photoelectrodes and a techno-economic analysis.



**Achievements in 2015:** DLR developed diverse reactor concepts, estimated related Ohmic losses using simplified models, and prepared a comprehensive assessment and optimization process. Reference experiments were successfully conducted and evaluated employing DLR's modified linear Fresnel concentrator SoCRatus (Solar Concentrator with a Rectangular Flat Focus), a two chamber suspension reactor, as well as particulate photocatalysts based on titanium dioxide ( $\text{TiO}_2$ ) and tin niobate ( $\text{SnNb}_2\text{O}_6$ ). Besides photocatalytic particles, water, methanol as a model substance for organic contaminants, and electrolytes formed part of the suspensions which received homogeneously concentrated solar irradiation in the focal plane of the concentrator with a geometric concentration ratio of 20.2 (Figure 4.10). Three temperature levels were considered: 25°C, 37.5°C, and 50°C. The production of hydrogen in the presence of methanol reached maximum values of 7386  $\mu\text{mol/h}$ . Despite of the larger band gap,  $\text{TiO}_2$  obtained clearly higher efficiencies than  $\text{SnNb}_2\text{O}_6$  with respect to the solar input in different spectral ranges.

**Publications:** [4.19]-[4.20]

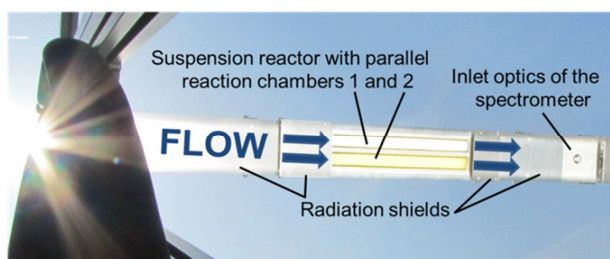


Figure 4.10. Irradiated suspension reactor in the focal plane of SoCRatus.

## HYDROSOL-PLANT – Thermochemical Hydrogen Production in a Solar Monolithic Reactor: Construction and Operation of a 1 MW Plant

**Participants:** APTL (GR), DLR (D), Hygear (NL), HELPRES (GR) and CIEMAT-PSA (E)

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**Funding:** FCH-JU-2012: € 2,500,000

**Duration:** January 1, 2014 - December 31, 2016

**Background:** The HYDROSOL-PLANT project is expected to develop, verify and operate all of the tools required to scale up solar  $\text{H}_2\text{O}$  splitting to the pilot (750  $\text{kW}_{\text{th}}$ ) scale. The work is based on the successful HYDROSOL series projects and mainly on the outcome of the current FCH-JU co-funded project, HYDROSOL-3D, dedicated to the provision of all main design specifications of such a pilot plant. HYDROSOL-PLANT

comes thus as the natural continuation of such an effort for  $\text{CO}_2$ -free hydrogen production in real scale.

**Purpose:** The main objectives of HYDROSOL-PLANT are to: (1) Define all key components and aspects necessary for the erection and operation of a 750  $\text{kW}_{\text{th}}$  solar plant for  $\text{H}_2\text{O}$  splitting (heliostat field, solar reactors, overall process monitoring and control, feedstock conditioning, etc.); (2) Develop tailored heliostat field technology (field layout, aiming strategies, monitoring and control software) that enables accurate temperature control of the solar reactors; (3) Scale-up the HYDROSOL reactor while advancing the state-of-the-art (redox materials, monolithic honeycomb fabrication and functionalization) for optimum hydrogen yield; (4) Design the overall chemical process, covering reactants and products conditioning, heat exchange/recovery, use of excess/waste heat, monitoring and control; (5) Construct a solar hydrogen production demonstration plant in the 750  $\text{kW}_{\text{th}}$  range to verify the developed technologies for solar  $\text{H}_2\text{O}$  splitting; (6) Operate the plant and demonstrate hydrogen production and storage on site (at levels > 3 kg/week) [4.21]; (7) Perform a detailed techno-economic study for the commercial exploitation of the solar process.

**Achievements in 2015:** The platform for hosting the HYDROSOL-Plant has been prepared by CIEMAT on the CRS-SSPS tower at the Plataforma Solar de Almería (PSA) and is ready for operation [4.21]. The receiver-reactor – as a fully assembled and fully functional system – will be mounted on the platform at a height of 28 m. An additional floor 4 m below will accommodate peripheral components.

Figure 4.11 (left) shows a picture of the refurbished experimental platform. The front has been designed to accommodate the three receivers according to the final configuration of the solar reactor array that is schematically depicted in Figure 4.11 (right). The solar chemical reactor consists of three cylindrical cavity-receivers containing a windowed aperture and a CPC, all arranged in an equilateral triangle. The distance of the receivers to the optical axis was determined based on the vertical space available on the tower and operation parameters like spillage losses and control temperature.



Figure 4.11. Picture of the refurbished experimental platform showing openings for the three receivers (left) according to the final configuration of the solar chemical reactor array schematically depicted (right).

In the previous plant investigated within the HYDROSOL-3D project, up to 70% of the incoming radiative power was lost due to thermal re-radiation at the hot absorber surface. Hence, the overall hydrogen production efficiency was reduced considerably. A preliminary simulation has been carried out and resulted in recommendations for getting an optimized homogeneous input flux providing low spillage losses.

Publication: [4.21]

## HITERSOL – Clean Technologies for Solar Hydrogen Production based on Mixed-Ferrites Thermochemical Cycle

Participants: ABENGOA Hydrogen € and CIEMAT-PSA (E)

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Funding: CTA-IDEA: € 150,000

Duration: June 1, 2014 – December 31, 2015

Background: HITERSOL is a cooperative project between CIEMAT-PSA and the company Abengoa Hydrogen, established within the framework of the CTA-IDEA Initiative funded by the Andalusia Community. Within a previous project named SolH2, a 200 kW<sub>th</sub> pilot plant was designed, constructed and commissioned on the CRS-SSPS tower at PSA. This installation is expected to serve as a platform for efficiently running thermochemical processes for the production of H<sub>2</sub>.

Purpose: The HITERSOL project aims to develop clean technologies for solar hydrogen production based on water splitting by mixed-ferrites thermochemical cycle. A challenging approach is pursued for the solar chemical reactor, which consists of a cavity receiver containing multiple tubular reactors.

Achievements in 2015: The present collaboration aims to complete the evaluation of the 200 kW<sub>th</sub> SOLH2 solar receiver that is located on the CRS-SSPS tower at 28 m height. It consists of a cavity receiver with 80 vertically oriented ceramic tubes containing commercial ferrite (Figure 4.12). The SolH2 facility also comprises auxiliary services, such as inert gas, water, compressed air and electricity supply, as well as communication wiring.

In 2015, CIEMAT carried out different activities of the project in collaboration with the ABENGOA Company, such as construction, commissioning and implementation of the software based on SCADA (System of Control, Supervision and Acquisition of Data). Preliminary simulations of the heliostat field including the heliostats required by the reactor design provided information on the feasibility of changing the heliostat reflection angle with sufficient speed and the realization of the necessary temperature levels in order to achieve



Figure 4.12. Photograph of the reactor for SolH2 tests on the CRS-SSPS tower at PSA.

the best performance (e.g. homogeneous radiative flux, low spillage losses, etc.).

Main conclusions of this study confirm that the solar power needed for the reaction is achievable and the proper flux distribution on the reaction chamber may become a limiting factor. At this stage, the definition of the heliostat aiming strategy has been completed and the operating conditions have been defined before the experimental chemical campaign scheduled for the beginning of 2016.

## ALCCONES – Storage and Conversion of Concentrated Solar Power

Participants: IMDEA Energía (E), Universidad J. Carlos I (E), ICP-CSIC (E), CIEMAT (E), SENER (E), ABENGOA Hydrogen (E)

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Funding: Community of Madrid: € 2,017,000

Duration: October 1, 2014 – October 31, 2017

Background: The ALCCONES project is a very ambitious initiative that involves the R&D public institutions IMDEA Energía (Coordinator), University J. Carlos I, ICP-CSIC and CIEMAT. SENER (a Spanish petroleum company) and ABENGOA Hydrogen (company subsidiary of the ABENGOA group) act as industrial companies with active collaboration and interest in the possible exploitation of the project results.

Purpose: The main goal of the program is to develop technologies for the conversion and storage at CSP systems for the efficient production of electricity, industrial process heat and hydrogen. The three objectives that provide functional structure to the work program of ALCCONES are: (1) Flowsheeting and multi-objective system analysis of CSP systems with integration of new thermal fluids; (2) develop new solar receivers and

reactors for the efficient operation at high temperatures and with high penetration of photons for high incident flux; (3) develop new storage and dispatching thermal energy systems such as: (a) thermocline beds, (b) encapsulated new PCM (Phase Change Materials), (c) thermochemical storage media with reversible reactions, (d) hydrogen produced by thermochemical cycles and stored in MOF (Metal Organic Framework) based materials for power, heat and hydrogen production on demand.

**Achievements in 2015:** Within objective (2), CIEMAT is exploring new solar receivers and reactors for the efficient operation at high temperatures and with high penetration of photons for high incident flux. In 2015, work was focused on studying the technical feasibility of chemical reactor concepts based on directly heated fluidized bed reactors. The fluidized reactor under consideration consists of a refractory lined steel cavity with a 200 mm diameter aperture. Commercial Ni ferrites have been selected as a model material for exploratory experiments in the fluidized bed reactor located on a solar parabolic dish. The test campaign was performed in continuous mode, filling the reactor with a controlled amount of granular material and  $N_2$  gas to ensure acceptable height of the moving bed and residence time. First experiments were focused on determining the best operating conditions, such as minimum gas velocity and height of the bed.

Within objective (3), research efforts by CIEMAT were directed towards the preparation and synthesis of perovskite materials – such as  $La_xSr_{1-x}Mn_yAl_{1-y}O_3$  and  $La_xSr_{1-x}Fe_yAl_{1-y}O_3$  – for two-step water splitting thermochemical cycles. The perovskites were tested in the laboratory under various reaction conditions with the goal to improve the kinetics and reduce the working temperatures. Comparability between different materials was established by TGA experiments, showing diverse weight losses for different samples investigated (Figure 4.13). First hydrolysis tests carried out with these perovskite materials indicate that the recovery of  $O_2$  is not complete within the operational conditions chosen.

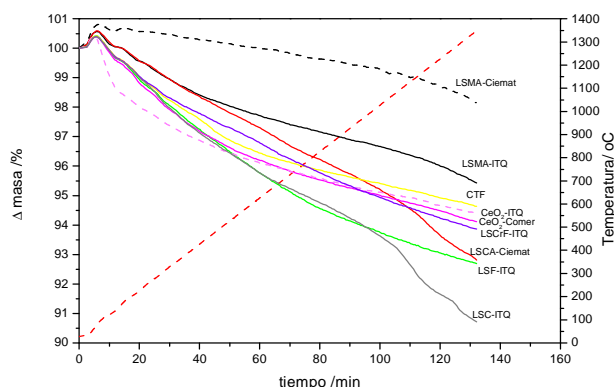


Figure 4.13. TGA experiments showing weight losses of various perovskite materials studied.

## ORESOL – Release of Oxygen from Lunar Regolith using Concentrated Solar Energy

**Participants:** CIEMAT-PSA (E)

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**Funding:** EC 6<sup>th</sup> Framework Programme ERA-STAR Regions (CA-515793- ERA-STAR REGIONS), ESP2007-29981-E.

**Duration:** September 1, 2014 – August 31, 2017

**Background:** The project ORESOL originates from the "ERA-STAR Regions" program (ERA-STAR – Space Technologies Applications & Research for Regions and Medium-Sized Countries), where it was part of a joint initiative between Andalusia/Spain and Bremen/Germany. PSA continued the activity due to the unique possibility to investigate a promising type of solar chemical reactor for reactions that require processing of large quantities of solids.

**Purpose:** The principal objective of the project is the development and testing of a solar powered fluidized bed reactor for the extraction of oxygen from lunar regolith. This is done by the reduction of one constituent of lunar soil, ilmenite ( $FeTiO_3$ ), with hydrogen, and the subsequent electrolysis of the obtained water.

**Achievements in 2015:** After finishing the assembly in 2014, the behavior of the solar reactor and the peripheral components was thoroughly tested under non-solar conditions. In particular, this included the operation of the reactor with the window installed to determine the pressure losses of all upstream and downstream elements. Finally, an important milestone was the relocation of the system from the laboratory (where the assembly and cold pre-tests were done) to the experimental platform of PSA's big Solar Furnace SF60. Presently, a major modification of this platform is under way. When finished, the solar test campaign can start.

Besides the work on the hardware of the reactor, a study was done with the goal to simplify the concentrator system for the operation on the lunar surface. While a conventional solar furnace (Figure 4.14 left) consists of a flat heliostat (1), a building for weather protection, an attenuator (2) to compensate for the varying solar radiation flux, the parabolic concentrator (3), the solar reactor (4) on an elevated working platform, and an additional water-cooled diagonal mirror (5) to achieve a vertical beam, the simplified and especially lightweight version (Figure 4.14 right) uses an off-axis concentrator that allows to leave the reactor on the ground. The hardware can be reduced to the flat heliostat (1), the off-axis concentrator (3), and the reactor (4). The performance, especially the achievable peak flux, is slightly reduced, but this is not relevant for this application. Due to its appearance, the concentrator was named "mussel-concentrator" [4.22].

A proposal called "SOLUNOX" has been submitted within the Horizon 2020 Call H2020-FETOPEN-2014-2015-RIA. SOLUNOX aims to set a solid scientific and technological basis for the first feasible approach of



oxygen ( $O_2$ ) production from lunar regolith. The target is to demonstrate the proof-of-principle sustainable operation of an integrated lunar  $O_2$  production plant towards the direction of Moon industrialization. This proposal has been prepared in collaboration with the APTL (Greece), DLR (Germany), Hygear (Netherlands) and CIEMAT-PSA (Spain).

**Publication:** [4.22]

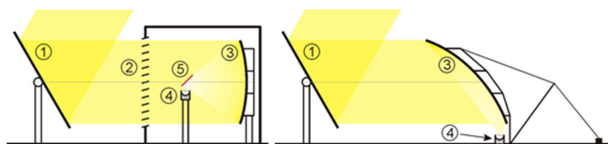


Figure 4.14. Conventional solar furnace (left). Off-axis or "mussel" concentrator (right).

## SEVR - Solar Expanding-Vortex Particle Receiver-Reactor Technology

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**Funding:** Australian Research Council (ARC) and Australian Renewable Energy Agency (ARENA)

**Duration:** January, 13, 2013 – February, 1, 2016

**Background:** The employment of directly-irradiated particle receivers for Concentrated Solar Thermal (CST) technology is gaining attention due to their potential to attain higher efficiency and higher temperatures than is possible with tubular receivers [4.23]. Of all the directly-irradiated particle receivers-reactors proposed to date, the Solar Vortex Reactor (SVR) has been found to be one of the most promising concepts, being applied successfully at laboratory scale to several applications, e.g. gasification of carbonaceous materials [4.24]. Furthermore, it has been found to have comparatively high energy conversion efficiency among several directly-irradiated solar reactors. However, despite its promising performance indicators, present SVR design has two drawbacks that pose a significant challenge to the robust operation of this receiver, namely a particle residence time distribution that is almost independent of particle size (meaning uneven heating of particles and exergy destruction) and the propensity to deposit particles onto the receiver-reactor window. These limitations adversely affect the energy efficiency and the chemical conversion, while also generating the risk of failure of the window. The University of Adelaide has patented a novel concept of solar vortex receiver-reactor for CST applications, termed the Solar Expanding-Vortex Particle Receiver-Reactor (SEVR) that addresses these challenges through a modification of the vortex structure. In

particular, the SEVR allows direct heat transfer whilst greatly mitigating any transport of flow through the aperture, to allow either the use of no window for some applications (notably where the atmosphere can tolerate a small air ingress), or the use of a very much lower purge flow to control particle deposition onto the window than has been used previously. Also, it achieves "aerodynamic classification" to control residence time distribution of the particles as a function of particle size and so to accomplish efficient processing of particles with a wide size distribution.

**Purpose:** The main objective is to develop and optimize the SEVR technology for different applications, including solar gasification of carbonaceous materials, mineral processing (alumina and/or magnesia production) and particle receiver. Numerical and experimental approaches will be employed to achieve the goal. Experimental validation of a 100 kW reactor model for heat/mass transfer and fluid flow will be performed.

**Achievements in 2015:** A schematic diagram of the SEVR configuration is reported in Figure 4.15, highlighting the key features of the SEVR, i.e. a conical expansion at the flow inlet, which is at the opposite end of the chamber to the aperture, and radial outlets. Promising experimental results were obtained in a 100 kW cold flow SEVR model. In detail, the first measurements of particle deposition onto the receiver-reactor window were carried out, showing that the particle deposition rate is reduced by  $> 10$  times in comparison with previous SVR design (Figure 4.16). The validated CFD model was employed to perform a systematic study of the effect of changing the geometric configuration of a 1 MW SEVR configuration on flow field, particle residence time distribution and particle deposition, under iso-thermal and non-isothermal conditions.

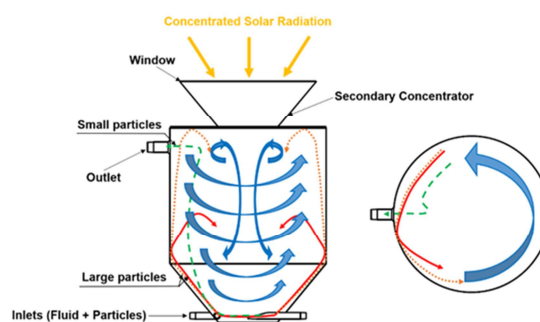


Figure 4.15. Schematic diagram of the Solar Expanding-Vortex Particle Receiver-Reactor (SEVR), showing a side view (left) and a bottom view (right), for the case with a vertical orientation. Also shown, a simplified representation of the three dimensional flow-field within the SEVR (blue lines), together with the key features of the deduced particle trajectories for the large (red), intermediate (orange) and small (green) particles to illustrate (left) the axial and radial components and (right) the tangential and radial components at the exit plane..



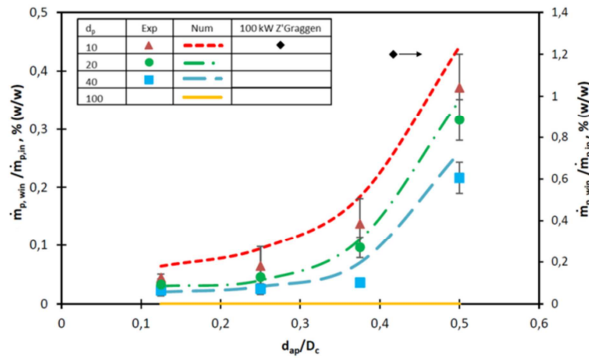


Figure 4.16. Measured and calculated total mass flow rate of particles deposited onto the receiver-reactor window relative to the total mass flow rate of inlet particles, as a function of normalized aperture diameter,  $d_{ap}/D_c$  ( $D_c$  is the chamber diameter), for a range of particles sizes,  $d_p$  ( $\mu\text{m}$ ), and a constant particle loading, for the 100 kW SEVR-40 configuration (owing to the  $40^\circ$  cone angle,  $\alpha$ ). The error bars correspond to one standard deviation in the data. Also shown is our calculated value for the 100 kW SVR Z'Graggen configuration [4.24], with the data on a different scale.

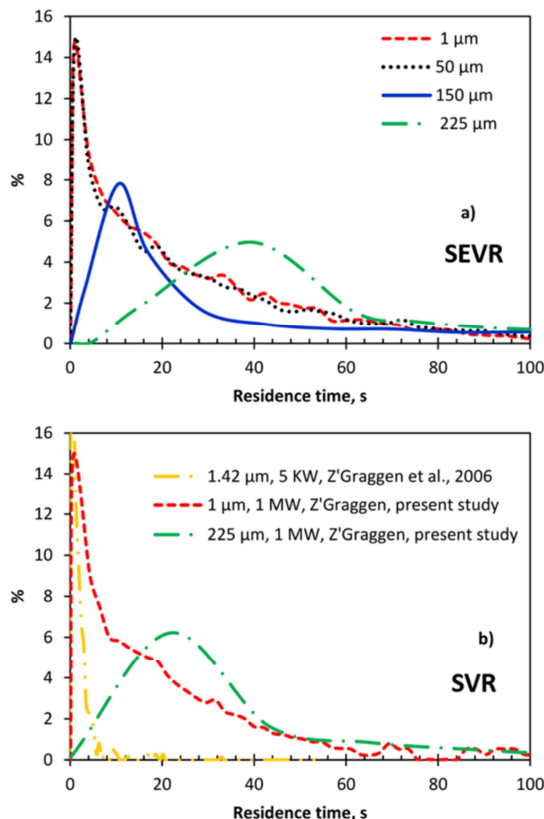


Figure 4.17. Particle residence time distribution relative to: (a) four different particle sizes (1, 50, 150 and 225  $\mu\text{m}$ ) for 1 MW configuration SEVR-50 (owing to the  $50^\circ$  cone angle), (b) two different particle sizes (1 and 225  $\mu\text{m}$ ) for the 'SVR Z'Graggen, 1MW' configuration, and 1.42  $\mu\text{m}$  for the 5 kW SVR of Z'Graggen et al. (2006) [4.24].

An aerodynamic mechanism with which to mitigate the particle deposition was identified as well as an optimized SEVR design. Furthermore, the residence time distribution of particles was calculated for different particle sizes, particle distributions (mono-sized or poly-disperse) and materials. Results showed that larger particles are preferentially recirculated within the chamber relative to smaller particles (Figure 4.17). Also, prolonged particle residence times can be achieved in comparison with the state-of-the-art of SVR technology.

Publication: [4.23]-[4.24]

## Hy-Sol-CLC – Hybrid Solar Chemical Looping Combustion with a Fixed Bed Reactor

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Funding: Australian Solar Thermal Research Initiative (ASTRI)

Duration: May, 31, 2015 – November, 31, 2015

Background: A chemical looping combustion (CLC) system is proposed to be hybridized with concentrated solar thermal energy (CST). This hybrid solar chemical looping combustion (Hy-Sol-CLC) system constitutes a further development of our earlier research to make it compatible with ASTRI's molten sodium receiver and also to avoid some of the technical challenges associated with implementing the vortex reactor as proposed earlier. In the new cycle, concentrated solar thermal radiation from the solar collector field is captured by the ASTRI solar cavity receiver to heat molten sodium, which is then used to provide the heat required within the fixed bed fuel reactor. The fixed bed is proposed to avoid the problems associated with the separation of gas and particles, and also to minimize the leakage of gas in the loop seals between the fuel and air reactors. In this concept, shown schematically in Figure 4.18, the solid oxygen carriers are kept stationary in a fixed bed reactor and are alternately exposed to reducing/oxidizing conditions through periodic switching of the fuel and air streams using valves 1 to 4. A configuration of shell and tube is proposed for the fuel reactor to heat the fixed bed of oxygen carriers using molten sodium, in which the hot molten sodium passes through the shell while the fixed bed oxygen carriers are in the tube. When the available concentrated solar thermal energy is sufficient to exceed the losses it is used to sensible heat the molten sodium. The produced hot molten salt is then applied to supply the sensible heating of the oxygen carrier bed and the heat required for the reduction of the bed with

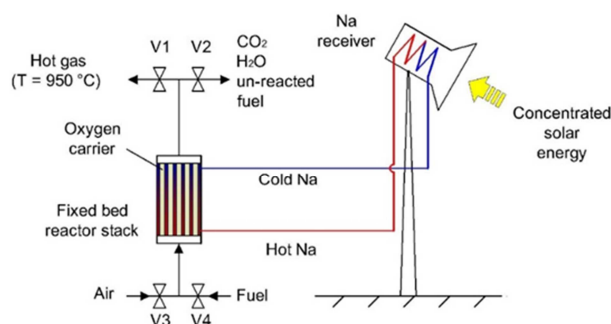


Figure 4.18. Schematic representation of a periodically operated packed bed solar hybrid chemical looping combustion. The valves 1-4 are proposed to periodically switch the fuel and air streams into the fixed bed oxygen carrier. The molten salt receiver together with is employed to provide the heat required within the fixed bed fuel reactor.

$\text{CH}_4$ , which is supplied through valves 4 and 2 ( $V_4$  &  $V_2$ ). The stored heat in the bed is then released through exothermic oxidation of the bed with air, using valves 1 and 3 ( $V_1$  &  $V_3$ ).

**Purpose:** The objective is to investigate the performance of this Hy-Sol-CLC system for the base-load power generation in terms of both the share of solar thermal energy in the system and coke formation on solid oxygen carriers in the operating temperature range of the ASTRI solar molten sodium receiver. Carbon formation on particles can decrease both the oxygen carrier bed reactivity and carbon capture efficiency of the system.

**Achievements in 2015:** In order to assess the kinetics of the reduction reactions of the oxygen carrier bed with  $\text{CH}_4$ , a 2-D unsteady-state model of a fixed bed reactor, operating in a reactor stack, has been developed. The model comprises the solution of conservation equations for energy, mass and momentum. The kinetic model of the reactions, as the key component of the model, has been solved. This section of the model has been also validated against the available experimental data in the literature. Figure 4.19 presents a comparison of the mole fraction of the gaseous components calculated by the model, with the experimental data reported by Iliuta

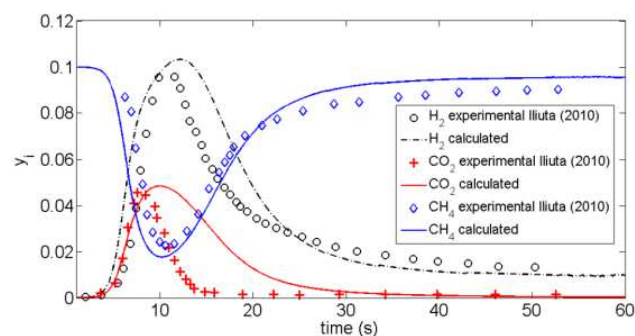


Figure 4.19. Comparison of the mole fraction of the  $\text{CH}_4$ ,  $\text{H}_2$  and  $\text{CO}_2$  in the outlet stream from the fixed bed reactor by the present model and the experimental data reported by Iliuta et al. (2010).

(2010)<sup>2</sup>. As can be seen good agreement was found between the model predictions and the experimental data.

**Publication:** [4.22]

## SCTL – Performance Assessment of Fischer-Tropsch Liquid Fuels Production by Solar Hybridized Dual Fluidized Bed Gasification of Lignite

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**Funding:** Australian Solar Thermal Research Initiative (ASTRI)

**Duration:** February, 3, 2014 – December, 31, 2016

**Background:** A dual fluidized bed (DFB) gasifier – an allo-thermal gasifier – is proposed to be hybridized with CST. In this system, the solid particle (bed material) is used as heat carrier to transfer the heat required by the gasification process from the combustion process and/or the solar receiver. Hence, the solar hybridized DFB (SDFB) gasifier offers the potential to store the solar thermal heat in the bed material as sensible heat in an additional storage tank. Furthermore, a steady syngas output can be achieved by maintaining the constant temperature of the hot bed material to the gasification process through adjusting the additional fuel input to the combustion process according to the solar radiation variation. However, no previous investigation has been reported of a hybrid DFB gasifier with CST.

**Purpose:** The objectives are (1) to compare the annual performance of a solar hybridized coal-to-liquids (SCTL) system including a SDFB gasifier with an equivalent non-solar CTL process in a way that accounts for solar variability using long-term historical measurements of direct normal irradiation (DNI), and (2) to assess the sensitivity of annual performance of the present studied SCTL system to the solar multiple, the storage capacity, the char conversion in the bubbling fluidized bed gasifier (BFBG) and the quality of the solar resource.

**Achievements in 2015:** This solar hybridized coal-to-liquids (SCTL) process is simulated using a pseudo-dynamic model that assumes steady state operation at each time step for a one-year, hourly integrated solar insolation time-series. The annual energetic and environmental performance of the SCTL system with a SDFB gasifier is analyzed as a function of solar multi-

<sup>2</sup> Iliuta I. et al. (2010). Chemical-looping combustion process: Kinetics and mathematical modeling. *AIChE J.* 56, 1063–1079.

ple ( $SM$ ), bed material storage capacity ( $SC$ ), char conversion in the BFBG ( $X_{char,BFBG}$ ) and the quality of solar resource. The  $SM$  is the ratio of the heliostat field area over the required heliostat field area which can generate net annual peak solar thermal equal to the heat required by the DFB gasifier. For the case of the minimum hot bed material storage capacity to avoid solar spillage ( $SC_{no-spill}$ ) and  $X_{char,BFBG} = 100\%$ , Figure 4.20 shows that the annual solar share ( $SS_{ann}$ ), the utilization factor of heliostat collector ( $U_{coll,ann}$ ), the percentage change in annual specific FTL output ( $\Delta Q_{s,FTL,ann}$ ) and annual reduction of  $CO_2$  emission ( $\Delta E_{CO_2,ann}$ ) relative to their non-solar CTL counterparts are increased to maximum values of 30.4%, 43.8%, 50.9% and 46.9%, respectively, as  $SM$  is increased to 4. However, the huge required  $SC$  (over 900 hours) impedes the feasibility of this case. Nevertheless, significant gains are also possible with more realistic  $SC$  even though some spillage results. For  $SM = 3$  and  $SC = 16$  hours,  $SS_{ann}$ ,  $U_{coll,ann}$ ,  $\Delta Q_{s,FTL,ann}$ , and  $\Delta E_{CO_2,ann}$  are calculated to be 21.8%, 43.1%, 32.6% and 34.1%, respectively. For the case of the presently assessed lignite fuel, the net mine-to-tank (MTT)  $CO_2$  emissions exceed the value of mineral crude. Nevertheless, this disadvantage can potentially be avoided or reduced by the use of low-net-carbon feedstocks, such as biomass and agricultural wastes.

Publication: [4.25]

## SCWG – Supercritical water gasification of algal biomass

Participants: The Australian National University (AUS), CSIRO (AUS), University of Adelaide (AUS), University of Colorado (USA)

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Funding: Australian Solar Thermal Research Initiative (ASTRI)

Duration: 2013–2016, possible extension to 2020

Background: Supercritical water gasification (SCWG) using solar thermal heat input is attractive as an option for the production of renewable liquid transport fuels from a range of carbonaceous feedstocks. Advantages of this concept include (1) high reaction rates, permitting compact reactor design, (2) gasification at lower temperatures (3) clean syngas with minimal  $NO_x/SO_x$  production, (3) efficient process for feedstocks with a wetness mass fraction  $>30\%$ , such as algae, bagasse and certain industrial wastes, (4) high heat input requirements make it attractive for optimization with an external heat supply, decreasing the consumption of the feedstock used for heating, (5) with biomass as the feedstock, the fuel product can be environmentally carbon-neutral, (6) minimal tar and char, (7) high carbon efficiency from feedstock to fuel appears feasible, (8) ash can be re-used as a fertilizer.

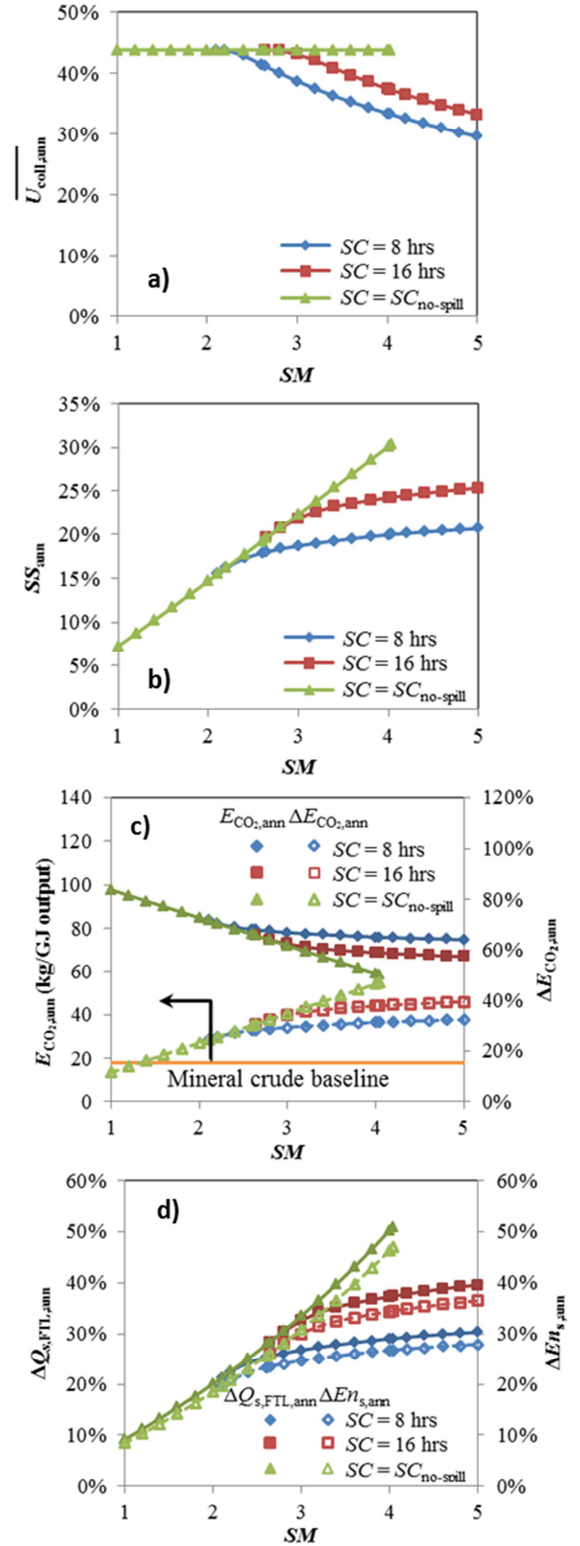


Figure 4.20. **a)** Annually averaged heliostat collector utilization factor ( $U_{coll,ann}$ ), **b)** Annual solar share ( $SS_{ann}$ ) of the SCTL system, **c)** Percentage change in annual specific FTL output,  $\Delta Q_{s,FTL,ann}$  (solid line) and specific total energetic output,  $\Delta En_{s,ann}$  (dash line) and **d)** Annual mine-to-tank (MTT)  $CO_2$  emissions,  $E_{CO_2,ann}$  (solid line) and annual reduction of MTT  $CO_2$  emissions,  $\Delta E_{CO_2,ann}$  (dash line) of the SCTL system relative to non-solar CTL system as a function of solar multiple ( $SM$ ) for different bed material storage capacities ( $SC$ ).



Challenges associated with SCWG include the high-pressure ( $>220$  bar), high-temperature ( $400\text{--}750^\circ\text{C}$ ) internal environment and the corrosive nature of reactants in supercritical conditions, as well as system-level challenges such as efficient thermal integration, especially the preheating and recovery of water used in the reactor, and carbon efficiency – turning as much as possible of the feedstock carbon into liquid fuel carbon, minimizing emissions and other wastes. Other key challenges are how to deal with process intermittency, especially noting the difficulties in operating Fischer-Tropsch fuel synthesis processes from a variable syngas source powered by sunlight.

This project aims to address these challenges, and to move SCWG solar fuels towards commercialization, while at the same time improving our understanding of the fundamental chemical and thermal processes taking place in these systems.

**Purpose:** The objectives are (1) to demonstrate experimental proof-of-concept for supercritical water gasification of algal biomass; (2) to develop a mechanistic understanding of the kinetics and thermodynamics in the SCWG process; (3) system modelling of the SCWG process and downstream integration with the Fischer-Tropsch, primarily targeting production of liquid transport fuel from biomass; (4) Techno-economic assessment of algae-to-fuels via SCWG/Fischer-Tropsch, targeting a 50% reduction in  $\text{CO}_2$  emissions (versus conventional), in a diesel fuel at a levelized cost below 2.50 AUD/L.

**Achievements in 2015:** We have developed a steady-state quasi-equilibrium reactor model for use in design of the lab-scale system, as well as for future use in modelling of solar-driven supercritical water gasification reactor. The chemical equilibrium in supercritical media is predicted using a Gibbs free energy minimization approach. The thermal and transport properties are calculated through modeling a real-gas mixture using Peng-Robinson equation-of-state and vdW mixing rules. The equilibrium model is coupled with standard heat transfer correlations for completing a 1-D model of the gasification reactor. Based on the understanding developed using the gasification reactor modeling, we have developed the design for a lab-scale standalone, continuous flow algae gasification reactor (Figure 4.21).

ANU has also been working on full-scale process simulation and techno-economics of an algae-to-fuels plant using AspenPLUS. Our efforts are focused on addressing prior limitations in carbon efficiency, heat utilization, phase separation problems and supercritical heat-exchange. Exergy efficiency methods have been used to identify the poorly performing parts of the process and strategies to circumvent these issues.

In parallel with the chemical process modeling in AspenPLUS, ANU and CSIRO are developing a new Modelica-based simulation framework for annual performance calculation of solar thermal energy systems. This framework is currently able to calculate the performance and economics of generic solar thermal plants, and has been validated against other models. The



Figure 4.21. Rendered layout of the SCWG experimental rig, currently under construction at ANU.

framework is now being adapted for the techno-economic assessment of solar thermochemistry processes such as SCWG/FT. The Modelica-based code is open-source, available at <https://github.com/SolarTherm>

**Publications:** [4.26]–[4.27]

### 4.3.2 ACTIVITY 6: MARKET PENETRATION (MP)

#### Roadmap to Solar Fuels – Strategy for Industry Involvement and Market Penetration

**Participants:** PSI (CH), DLR (D), WIS (IL), CSIRO (AUS), NWU (RSA), CSIR (RSA)

**Contact:** Anton Meier, [anton.meier@psi.ch](mailto:anton.meier@psi.ch)

**Funding:** SolarPACES ExCo funded project, cost shared: €25,000

**Duration:** July 1, 2012 - October 31, 2016

**Background:** This SolarPACES Task II Special Activity aims at enhancing industry involvement in solar fuels production and promoting market penetration of solar fuels technologies. Initially, two sun-rich countries—Australia and South Africa—expressed their keen interest to organize workshops with SolarPACES experts presenting to targeted local industry and governments the state-of-the-art and the market potential of the most advanced technologies for solar fuels production. The final outcome will be a country-specific “Roadmap to Solar Fuels” being initiated together with the identified industries and other interested bodies. If successful, such missions may later be extended to other countries involving different industries and various solar fuels or materials production technologies.



SolarPACES has the expertise in developing the solar technology for virtually any process producing solar fuels ( $H_2$ , syngas, or liquid fuels such as methanol, diesel, and jet fuel) and chemical or material commodities (metals, lime, and cement), as well as in providing solutions for thermo-chemical storage. It is important to spread this message among industry and governments.

**Purpose:** A “Roadmap to Solar Fuels” shall be developed with the objective of facilitating industry involvement and market penetration. In a first step, potential industrial and governmental players in two selected countries—Australia and South Africa—shall be identified and contacted. First-hand information on  $CO_2$  mitigation and market potential of solar fuels technologies shall be presented by SolarPACES experts at specific workshops and meetings organized by the corresponding Task II national coordinators (NCs). In a second step, a specific roadmap concept shall be initiated and tailored for each country, showing the most promising options for solar fuels as a result of a screening analysis by the local and external SolarPACES experts, interested industries, and responsible governmental representatives.

**Achievements:** In Phase 1 (2012-2013) of this special Task II activity on solar thermal fuels, the state-of-the-art and the market potential of the most advanced technologies for solar fuels production was presented by external SolarPACES experts to targeted local industry and governments in Australia and South Africa.

Major outcome of Phase 1:

- It was obvious that many attendees of the workshops were not aware of the work being done worldwide on solar thermal fuels. Thus, there is a need to increase the awareness of solar thermal fuels, to evaluate the previous R&D work done in this field, and to determine the applicability and feasibility of specific solar fuels technologies in countries like Australia and South Africa<sup>3</sup>, as well as China and many others. Furthermore, it is important to emphasize the economic and political aspects of solar fuels that could be a major export opportunity, as well as providing security of supply.
- Although the transition to a hydrogen economy seems remote for countries such as Australia, South Africa, or China, hydrogen is consumed in large quantities for fertilizer manufacture and refining. Therefore, solar production of hydrogen using thermochemical cycles based on metal oxide redox reactions should be considered as *long-term option* to produce fully renewable solar fuels.

- In the *near term*, the focus should be on syngas, from which one can make a wide variety of synthetic liquid fuels such as methanol, diesel, and jet fuel. Syngas can be produced from steam-reforming of natural gas (NG), oil, and other hydrocarbons, and steam-gasification of solid carbonaceous feedstock such as coal, coke, biomass, bitumen, and carbon-containing wastes. Solar reforming of NG, using either steam or  $CO_2$ , and solar-driven gasification have been extensively studied in solar concentrating facilities with small-scale solar reactor prototypes. The first generation of industrial solar reforming and gasification pilot plants using solar tower concentrating systems is coming into operation. Ultimately, solar reforming and gasification are an efficient means of storing intermittent solar energy in a transportable and dispatchable chemical form.
- Solar gasification of carbonaceous feedstock is particularly suited for sun-rich countries with abundant domestic coal/coke reserves such as Australia, South Africa, and China. Australia has also large reserves of natural gas but no liquid fuel and, therefore, solar reforming could be an attractive option there. In the end, successful market entry of solar reforming and gasification technologies will depend on the dominant market price of solid feedstock and fossil fuels, as well as on credits for pollution abatement and  $CO_2$  mitigation.

In Phase 2 (2013-2014; extended until 2015), progress in developing a solar fuels roadmap was made in both selected host countries (Australia and South Africa). Contacts with potentially interested industries and responsible governmental representatives have been established. Opportunities for joint projects involving industry, government agencies and international solar research centers have been identified and some of them are expected to be concretized in the near future.

Major outcome of Phase 2:

- Screening of potential solar fuels processes and a techno-economic evaluation of CSF technologies eventually led to country-specific roadmaps to solar fuels for Australia and South Africa.
- This study published on the SolarPACES Task II website [4.28] is a role model for broadening the effort, offering finally the whole SolarPACES community to benefit from the country-specific roadmaps and the methodology developed.
- The outcome of the roadmaps shall enable the participating countries to accelerate the implementation of solar fuel production as well as a methodology that makes this attempt transferable to other SolarPACES members, or to countries that are of special interest for the SolarPACES members but are not members by themselves yet. The project and the roadmaps for the participating countries shall be connected to existing roadmaps like the one of the European Industrial Initiative on solar energy – CSPi – and national roadmaps and political guidelines. An example is the solar hybrid roadmap of the Australian

<sup>3</sup> The SolarPACES “Roadmap to Solar Fuels” activity is providing additional international support to the Australian solar research community in developing their own “Solar Hybrid Fuels” roadmap for Australia funded by the Australian Renewable Energy Agency (ARENA). Another goal of the SolarPACES roadmap exercise is to help securing acceptance of solar thermal fuels to be included in the Solar Energy Technology Road Map (SETRM) for South Africa, thus providing a potential funding opportunity for the solar research community and industry in RSA.

an Renewable Energy Agency ARENA [4.29]. There shall be a link between the existing Concentrating Solar Power (CSP) industry and the future Concentrating Solar Fuel (CSF) industry as it will most probably integrate stakeholders from the CSP side as well as from the oil and gas companies and the utilities. Therefore, the goal is to show where early market entries might be feasible, especially focused on unique conditions in the SolarPACES member countries.

In Phase 3 (2015-2016), a solar fuels roadmap – similar to that developed for Australia and South Africa – has been initiated for China. Workshops/conferences and meetings will be held in China for the preparation of a country-specific roadmap concept, showing the most promising options for solar fuels as a result of a screening analysis by the local and external SolarPACES experts, identified industries, responsible governmental representatives and other interested bodies.

Major outcome of Phase 3 (ongoing):

- In December 2015, an exploratory workshop has been held in Xi'an, China, with solar technology overview presentations given by SolarPACES experts (Michael Epstein, Anton Meier, and Christian Sattler). The workshop was well organized and attended. The audience included 14 participants (mostly professors) from eight Chinese universities, about 30 scientists, postdocs and PhD students from Xi'an Jiaotong University, and four participants from industry.
- Other than South Africa and Australia, China is a net importer of coal and the largest user of coal in the world. The passing of the Renewable Energy Law (REL) in 2005 demonstrated China's commitment to renewable energy development<sup>4</sup>. Biomass had a significant role for fuel production in the REL. However, as the development of renewable energy applications continues, China is also developing alternatives, especially because the potential of biomass is rather limited.
- The third phase of the SolarPACES solar fuels roadmap project will contribute to this development by supporting to raise the awareness of concentrating solar technologies, inform stakeholders from academia, industry and policy on the state of the art and the perspectives, and develop a roadmap describing possible actions to intensify the effort to be put into solar thermochemical fuels.
- Figure 4.22 shows the current status of this ongoing Task II activity. Actions completed – such as the workshop in December 2016 – are highlighted in green. Activities planned for the first half of 2016 (highlighted in yellow) – such as preparing the initial roadmap concept and scheduling intended visits to high-level decision makers – are progressing slowly and will require special effort from the Chinese side. Overall, this leads to a delay in preparing the final roadmap document, which in turn is a prerequisite for political actions that aim to push the solar fuels roadmap becoming official policy in China.

Date	Activity	Deliverable	Responsible
September 2014	Task II Meeting in Beijing	Critical review of Phase 2 and preliminary planning of Phase 3	Task II OA NC CHN
Jan - June 2015	Identification and invitation of stakeholders	List of potentially interested industries and government departments	NC CHN
December 2015 (delayed from August 2015)	Workshop with technical sessions; Education of industry and gov. reps	Participants list; Meeting minutes; List of topics/challenges for "Roadmap to Solar Fuels"	NC CHN with support of SP experts
Jan – April 2016	Preparation of initial roadmap	Draft of "Roadmap to Solar Fuels" document; Intermediate report	NC CHN and SP experts
May / June 2016	Targeted visits to high-level decision makers; Improve roadmap	Participants list; Meeting minutes; List of topics/challenges for "Roadmap to Solar Fuels"	NC CHN with support of SP experts
July – Sep 2016	Push roadmap to be national policy in China	Final "Roadmap to Solar Fuels" document; Final report to ExCo	Task II OA NC CHN
October 2016	Task II Meeting in Abu Dhabi, UAE	Critical review of Phase 3	Task II OA NC CHN

Figure 4.22. Time schedule of "Roadmap to Solar Fuels" (Phase 3) showing activities, deliverables, and responsible persons.

<sup>4</sup> Feng Wang, Haitao Yin, Shoude Li, China's renewable energy policy: Commitments and challenges, Energy Policy 38 (2010) 1872–1878, doi:[10.1016/j.enpol.2009.11.065](https://doi.org/10.1016/j.enpol.2009.11.065)

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